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the Total Environment  
Manuscript Draft

Manuscript Number: STOTEN-D-18-06694

Title: Evaluation of four modelling approaches to simulate nitrous oxide emissions in China's cropland

Article Type: Research Paper

Keywords: nitrous oxide; model simulation; cropland; DAYCENT; DNDC; linear model

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**Abstract:** Process-based models are useful tools to integrate the effects of detailed agricultural practices, soil characteristics, mass balance, and climate change on soil N<sub>2</sub>O emissions in soil - plant ecosystems, whereas static, seasonal or annual models often exist to estimate cumulative N<sub>2</sub>O emissions under data-limited conditions. A study was carried out to compare the capability of four models to estimate seasonal cumulative fluxes from 425 field measurements of N<sub>2</sub>O emissions representing 67 studies across China's croplands. The models were 1) the DAYCENT model, 2) DeNitrification - DeComposition model (DNDC), 3) the linear regression model (LRM) of Yue et al. (2018), and 4) IPCC Tier 1 emission factors. The DAYCENT and DNDC models were estimated crop yields with R<sup>2</sup> values of 0.60 and 0.66 respectively; but DNDC showed significant underestimation according to bias analysis. For seasonal cumulative N<sub>2</sub>O emission predictions, the correlation of modelled with measured N<sub>2</sub>O emissions had an R<sup>2</sup> of 0.14, 0.14, 0.23 and 0.15 for DAYCENT, DNDC, LRM of Yue et al. (2018), and IPCC, respectively. No significant bias was identified except for the significant underestimation of 0.52 kg N<sub>2</sub>O-N ha<sup>-1</sup> with the DNDC model. The modelled daily N<sub>2</sub>O emission against observations from the experimental fields indicated that the DAYCENT and DNDC models simulated temporal patterns effectively, although they did not capture the emission peaks perfectly. Based on RMSE and bias analysis, LRM performed well on N<sub>2</sub>O emission prediction for paddy rice fields, while DAYCENT performed well for wheat and IPCC for maize. All models simulated N<sub>2</sub>O fluxes well for soybeans, but not well for cotton or fallow. Moreover, DAYCENT and LRM performed well under different fertilizer management (no fertilizer, mineral fertilizer, and organic fertilizer), while DNDC significantly underestimated the emissions under no fertilizer and when organic fertilizer was applied, as did IPCC when organic fertilizer was applied.

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Dear editors,

Herewith we are submitting a manuscript entitled “Evaluation of four modelling approaches to simulate nitrous oxide emissions in China’s cropland”, for review and potential publication in Science of the Total Environment. This manuscript provides an evaluation of four modelling approaches to simulate N<sub>2</sub>O emissions with 425 field measurements from China. We conclude that neither of the models emerged as a clear “best” choice for estimating N<sub>2</sub>O emissions for Chinese cropping systems.

This work has not been submitted or published elsewhere. The manuscript deals with the true results based on a newly established dataset of data collected from published literatures.

Please contact me if you have questions about the manuscript. We appreciate any consideration given to this manuscript for publication in Science of the Total Environment.

Sincerely,

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# Evaluation of four modelling approaches to simulate nitrous oxide emissions in China's cropland

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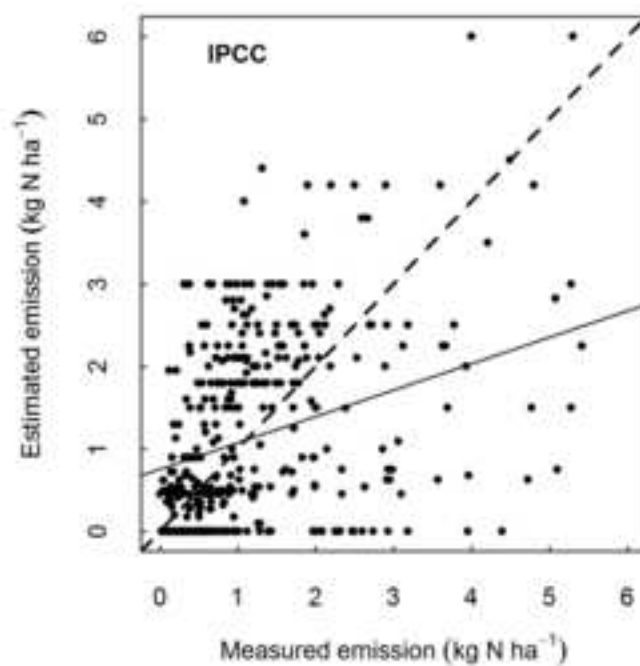
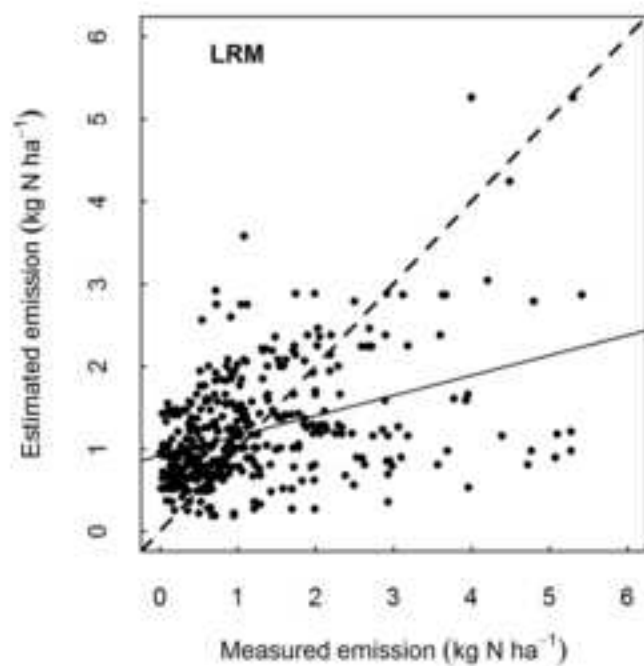
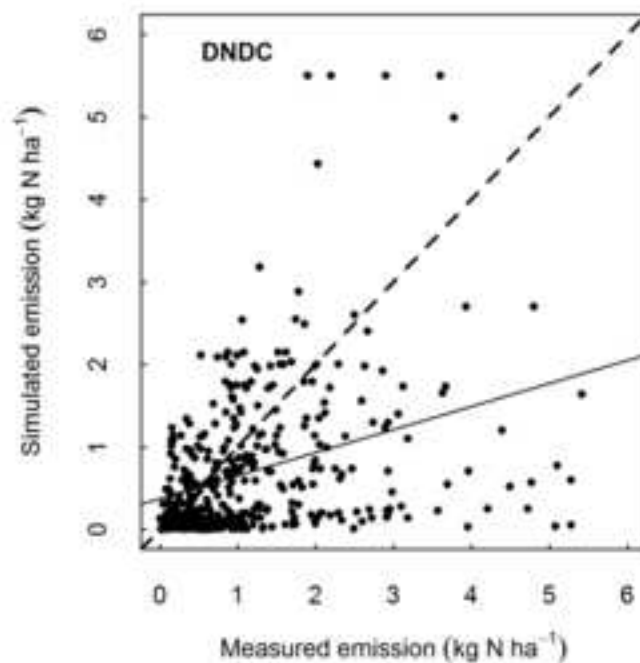
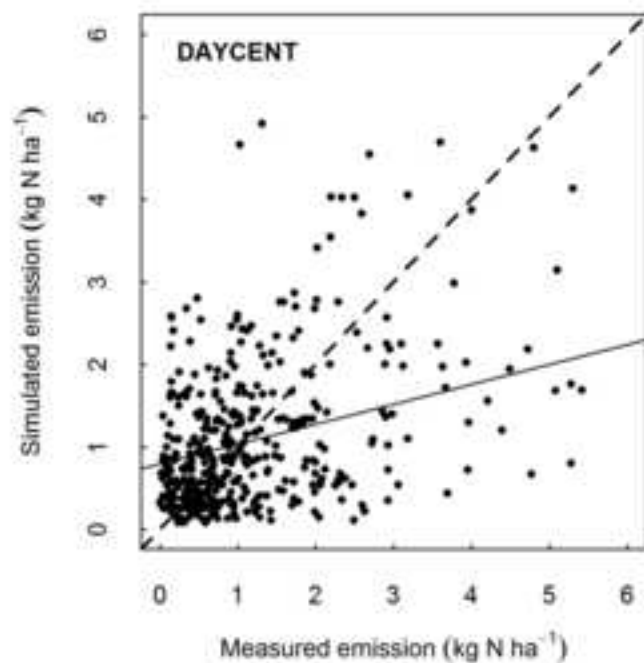
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Running head: Model comparisons for soil N<sub>2</sub>O emissions



## Highlights

1, DAYCENT, DNDC, linear regression model and IPCC tier 1 approach were evaluated to simulate N<sub>2</sub>O emissions in China's cropland.

2, Neither of the models emerged as a clear "best" choice for estimating N<sub>2</sub>O emissions for Chinese cropping systems.

3, Further development is needed to represent regional conditions in China.

# Evaluation of four modelling approaches to simulate nitrous oxide emissions in China's cropland

**Abstract:** Process-based models are useful tools to integrate the effects of detailed agricultural practices, soil characteristics, mass balance, and climate change on soil N<sub>2</sub>O emissions in soil - plant ecosystems, whereas static, seasonal or annual models often exist to estimate cumulative N<sub>2</sub>O emissions under data-limited conditions. A study was carried out to compare the capability of four models to estimate seasonal cumulative fluxes from 425 field measurements of N<sub>2</sub>O emissions representing 67 studies across China's croplands. The models were 1) the daily time-step version of CENTURY (DAYCENT), 2) DeNitrification - DeComposition model (DNDC), 3) the linear regression model (LRM) of Yue et al. (2018), and 4) IPCC Tier 1 emission factors. The DAYCENT and DNDC models were estimated crop yields with R<sup>2</sup> values of 0.60 and 0.66 respectively; but DNDC showed significant underestimation according to bias analysis. For seasonal cumulative N<sub>2</sub>O emission predictions, the correlation of modelled with measured N<sub>2</sub>O emissions had an R<sup>2</sup> of 0.14, 0.14, 0.23 and 0.15 for DAYCENT, DNDC, LRM of Yue et al. (2018), and IPCC, respectively. No significant bias was identified except for the significant underestimation of 0.52 kg N<sub>2</sub>O-N ha<sup>-1</sup> with the DNDC model. The modelled daily N<sub>2</sub>O emission against observations from the experimental fields indicated that the DAYCENT and DNDC models simulated temporal patterns effectively, although they did not capture the emission peaks perfectly. Based on RMSE and bias analysis, LRM performed well on N<sub>2</sub>O emission prediction for paddy rice fields, while DAYCENT performed well for wheat and IPCC for maize. All models simulated N<sub>2</sub>O fluxes well for soybeans, but not well for cotton or fallow. Moreover, DAYCENT and LRM performed well under different fertilizer management (no fertilizer, mineral fertilizer, and organic fertilizer), while DNDC significantly underestimated the

25 emissions under no fertilizer and when organic fertilizer was applied, as did IPCC when  
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## 1 Introduction

Anthropogenic greenhouse gas (GHG) emissions, a major contributor to climate change (IPCC, 2013), have increased rapidly across the world by 41% from 38.2 Pg CO<sub>2</sub> equivalent (CO<sub>2</sub>-eq) in 1990 to 53.9 Pg CO<sub>2</sub>-eq in 2012 (<http://edgar.jrc.ec.europa.eu/>). The trend is projected to continue in coming decades as a result of increasing food demand and limited resources (Reay et al., 2012). Meanwhile, the Paris Agreement aims to limit global warming to “well below” 2 degrees Celsius, with an ambition to pursue efforts to limit warming to below 1.5 degrees Celsius, and many countries have already made commitments to participate towards achieving these goals. As one of the world's most populous countries, with 29.3% of the world's total emissions (Janssens-Maenhout, et al., 2017), China is of key importance for mitigating global emissions, and has recently pledged “no-increase” in chemical fertilizer and pesticide in order to achieve peak GHG emissions by the year 2030 (UNFCCC, 2015).

Nitrous oxide (N<sub>2</sub>O) has a global warming potential (GWP) of approximately 265-310 times that of carbon dioxide (CO<sub>2</sub>) over a 100-year timescale (Watson et al., 1996; IPCC, 2007; IPCC, 2013) with an atmospheric lifetime of approximately 120 years (Prather, 1998). Global N<sub>2</sub>O emissions increased to 9.2 Tg N<sub>2</sub>O in 2012 from 5.4 Tg N<sub>2</sub>O in 1970. N<sub>2</sub>O contributes to secondary inorganic aerosol formation and thus haze pollution in addition to climate change (Liu et al., 2017). For China, N<sub>2</sub>O emissions accounted for 16.4% of global emissions (Janssens-Maenhout, et al., 2017). The most significant source of N<sub>2</sub>O emissions was agriculture, accounting for 51% of total national N<sub>2</sub>O emissions (FAO, 2015). Emissions from agriculture tripled from 0.36 to 1.21 Tg N<sub>2</sub>O in China between 1970 and 2014 (FAO, 2015). Given the importance of this source of emissions, reducing uncertainty in its estimation is an important issue for China to effectively identify ways to mitigate.

The Intergovernmental Panel on Climate Change (IPCC) provided in 1997 (IPCC, 1997) a default global N<sub>2</sub>O emission factor intended for use in national inventories of 1.25% with the confidence interval of 0.25-2.25% for fertilizer-induced emission (FIE) from all cropland (IPCC, 1997). That is, that 1.25% of nitrogen applied in crop systems is released as N<sub>2</sub>O-N. This factor was subsequently updated to 1% with the confidence interval of 0.3-3.0% and 0.3% with the confidence interval of 0.0-0.6% (Tier I approach) from upland crops and paddy rice cultivation, respectively (IPCC, 2006). Generally, the emission factor approach makes it easy to calculate the FIE using applied N rate, but also leads to large uncertainties. Therefore, as recommended by the IPCC (2006), higher Tier methods should be developed to obtain more representative, country specific emission rates or spatially disaggregated N<sub>2</sub>O-EFs that are region and crop-specific.

Linear or nonlinear regression models can be developed to estimate N<sub>2</sub>O emissions from croplands as a function of field and management variables based on field measurements (Bouwman et al., 2002; Gerber et al., 2016; Albanito et al., 2017). For example, Yue et al. (2018) published a China-specific multi-variate empirical model for N<sub>2</sub>O emissions to identify the spatial variability caused by the major drivers. On the other hand, process-based models have been widely used to estimate N<sub>2</sub>O emissions and potential effects of global climate change on the terrestrial ecosystems. Several dynamic process-based models have been developed to predict N<sub>2</sub>O emissions informed by an understanding of key soil processes and mechanisms (e.g. SUNDIAL by Smith et al., 1997; DNDC by Li et al., 1992; DAYCENT by Ogle et al., 2010). Compared to regression models, most process-based models simulate the emissions of several GHGs (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O) considering environmental and management related factors, such as crop growth, soil properties, fertilization and climate. DAYCENT and DNDC models are both widely-used ecosystem biogeochemistry models adopted to simulate N<sub>2</sub>O emissions all over the world (Abdalla et al., 2010). DAYCENT

77 simulates C, N, P, K and S dynamics in plant-soil systems (Parton et al., 1998; Del Grosso et  
78 al., 2001). DNDC mainly focuses on nitrification and denitrification for N dynamics from  
79 upland soils and rice paddy systems (Li et al., 1992, 1994).

80 There are limitations and uncertainties in estimating N<sub>2</sub>O fluxes from process-based model  
81 simulations, associated with the representation of the mechanistic processes. Frolking et al.  
82 (1998) found that DNDC simulated very low N<sub>2</sub>O fluxes for a dry site in Colorado. In  
83 contrast, Smith et al. (2007) produced accurate predictions of average seasonal N<sub>2</sub>O  
84 emissions from the DNDC model for two sites in Eastern Canada, while the DAYCENT  
85 model underestimated N<sub>2</sub>O emissions. This variability in performance implies that model  
86 inter-comparisons are useful to determine the most appropriate application for a specific  
87 region or cropping system. For many countries, including China, model inter-comparisons are  
88 especially important since many process-based models, in spite of their intent to be generic,  
89 were originally calibrated on data from North-American or European cropping systems. The  
90 objective of this study is to compare the results of four models, namely DAYCENT, DNDC,  
91 LRM, IPCC, by calibrating and evaluating the N<sub>2</sub>O emission estimates under different  
92 cropping systems and N application rates across the major agricultural regions of China.

## 2 Materials and methods

### 2.1 Model descriptions

DAYCENT, the daily time-step version of CENTURY, is a process-based ecosystem model developed to simulate carbon (C), N, P, K and S dynamics in plant-soil systems (Parton et al., 1998; Del Grosso et al., 2001). The nitrogen fluxes through the plant, residue and soil organic matter pools are coupled with C and estimated based on the C transfer between conceptual soil C pools, and the C:N ratio of organic matter. The model considers symbiotic and asymbiotic N fixation, and fertilizer additions. Losses of N occur through removal of vegetation, nitrification, denitrification,  $\text{NH}_3$  volatilization, leaching and run-off. Daily weather data, essential management events, and soil texture data are needed as model inputs (Table 1). For our study, historical runs were performed to initialize the model in accordance with China-specific conditions (details are described in Cheng et al. (2014)).

The DeNitrification - DeComposition model (DNDC), contains four main sub-models as follows: the soil climate sub-model calculating hourly and daily soil temperature and moisture fluxes in one dimension; the crop growth sub-model simulating crop biomass accumulation and partitioning; the decomposition sub-model calculates decomposition, nitrification,  $\text{NH}_3$  volatilization and  $\text{CO}_2$  production; and the denitrification sub-model tracking the sequential biochemical reduction from nitrate ( $\text{NO}_3$ ) to  $\text{NO}_2^-$ , NO,  $\text{N}_2\text{O}$  and  $\text{N}_2$  (Li et al., 1992; Li et al., 2000; Abdalla et al., 2010). Version 9.5 of the DNDC model was applied in the present study (<http://www.dndc.sr.unh.edu/>). The input data required were the same as for DAYCENT (Table 1).

A linear regression model approach has also been applied, named as LRM, fitting cumulative  $\text{N}_2\text{O}$  emissions (*Cum N<sub>2</sub>O*) in  $\text{kg N ha}^{-1} \text{ season}^{-1}$  based on the following equation:

$$Cum N_2O = Exp(-2.709 + 0.004 \times N\ rate + 0.074 \times Temp + 0.013 \times Clay + \beta_1 \times crop\ type + \beta_2 \times N\ rate \otimes fert\ type + \varepsilon)$$

where *N rate* represents the nitrogen fertilizer application in kg N ha<sup>-1</sup>; *Temp* is the annual average temperature (°C); *Clay* indicates the fraction of clay (%); values of  $\beta_1$  for the different crop type classes are 0 for legume, 0.700 for upland crops, -0.188 for rice; and values of  $\beta_2$  for the different base fertilizer are 0 for mineral fertilizer and -0.002 for organic fertilizer, and 0 for no fertilizer applied. The required data are N fertilizer application rates, annual average temperature, soil clay content, crop type, and fertilizer type (Table 1).

Finally, using the IPCC default method (2006), annual cumulative N<sub>2</sub>O emissions (*Cum N<sub>2</sub>O*) in kg N ha<sup>-1</sup> year<sup>-1</sup> are calculated using the following equation:

$$Cum N_2O = N\ rate * EF$$

where *N rate* represents the nitrogen fertilizer application in kg N ha<sup>-1</sup>; and values of *EF* are 0.01 and 0.003 for upland crops and paddy rice cultivation, respectively. The only required data are N fertilizer rates (Table 1).

## 2.2 Data sources

N<sub>2</sub>O emissions data were collected during the crop growing season at the experimental sites (kg N ha<sup>-1</sup> season<sup>-1</sup>) - defined as the period from planting to harvest for a given crop. We conducted a literature search in the databases: CNKI, ISI-Web of Knowledge and Google Scholar, with the search words “nitrous oxide”, “emission”, “chamber”, and “China”. A total of 134 papers were found and processed according to the publication date, journal category and data integrity. For these papers, a dataset of 67 studies with a total of 425 field N<sub>2</sub>O emission measurements were compiled. The dataset included the following information: cumulative N<sub>2</sub>O-N emissions; grain yields; geographic information; soil characteristics including clay content, C and N content, bulk density, and pH; cropping system; management

practices; crop types - maize (MA), wheat (WH), rice paddy (RP), soybean, cotton, rape, and fallow; and fertilizer types classified into 5 broad categories - Control, Mineral, Organic, Mineral & Organic (M\_O), Controlled-release fertilizer or Nitrification inhibitor (more detailed information is provided in Table S1). All data were used to test DAYCENT, DNDC, and the EF method of IPCC. It should be noted that 267 N<sub>2</sub>O field emission measurements of the whole database (425 measurements) were used to derive the linear regression model of Qian et al. (2018), with all the measurements used to test the LRM of Qian et al. (2018), so the LRM model is not entirely independent of the evaluation data.

Most of the soil, crop, and cultivation management data were obtained from the dataset. However, missing soils data that were not provided in the papers were extracted from China Soil Scientific Database (<http://www.soil.csdb.cn/>) based on the soil type documented for the experimental site. Similarly, missing daily weather data, including daily maximum/minimum temperature and precipitation, were obtained from the China Meteorological Data Sharing Service System (<http://new-cdc.cma.gov.cn:8081/home.do>) for the station nearest to the reported site. Regional nitrogen deposition data were based on Xu et al. (2015).

For the process-based models, most of the parameters were based on prior research (DAYCENT with information from Cheng et al., 2014; DNDC from Abdalla et al., 2010). Crop growth directly controls soil water and C and N regimes, and hence is crucial for a biogeochemical model to correctly simulate trace gas fluxes, such as N<sub>2</sub>O (Hu et al., 2017). PRDX (the maximum potential production parameter), a dimensionless constant, was optimized by simulating crop yields in the range of 1-3 for DAYCENT for each of the experimental sites. Similarly, the indices of maximum biomass production, biomass fraction, and biomass C/N ratio of grain, leaf, stem, and root distributions were optimized for yield simulations of field conditions for DNDC.

## 2.3 Model validation

### 2.3.1 Daily emission evaluation

DAYCENT and DNDC simulation results were evaluated against field measurements of N<sub>2</sub>O emissions by comparing the association between measured and modelled temporal patterns of N<sub>2</sub>O fluxes, as well as comparing the coincidence between measured and modelled emission values. Five representative benchmark sites were selected from the major regions of China to conduct the model evaluation of daily N<sub>2</sub>O emissions under typical cropping systems (Table 3). Daily measured emission values for model evaluation were extracted either directly from tables or text, or were extracted from the figures using Getdata Graph Digitizer software (<http://www.getdata-graph-digitizer.com/>).

### 2.3.2 Model accuracy

Cumulative N<sub>2</sub>O-N fluxes were estimated as the sum of simulated daily fluxes for DAYCENT and DNDC models, and directly by LRM and IPCC. Model accuracy was evaluated by calculating the bias and root-mean-squared error (RMSE) between measured and modelled values using the following equations:

$$Bias = \sum_{i=1}^n (\hat{V}_i - V_i) / n$$

$$RMSE = \sqrt{(\sum_{i=1}^n (\hat{V}_i - V_i)^2) / n}$$

where,  $\hat{V}_i$  and  $V_i$  represent the estimated value of the target variable from the fitted equation and the measured value from the original studies;  $\bar{V}$  is the mean of the observed data;  $n$  is the number of target values;  $p$  is the number of parameters in the relevant model; and  $i$  is a single observation.

With the estimated *Bias*, the t-statistic was used to test for significant differences between simulated values and measurements (Smith et al., 1997). *Bias*, *RMSE*, and a t-test statistic

188 were also calculated to evaluate model performance for each fertilizer and crop type  
189 individually. All the statistical analyses were conducted in R version 3.4.0 (2018) and  
190 Microsoft Excel 2013.

191



## 3 Results

### 3.1 Yield simulation

The measured yield data included 283 individual observations (Fig. 1), which ranged from 400 to 15700 kg ha<sup>-1</sup>. The modelled yields ranged from 537 to 16657 kg ha<sup>-1</sup> for DAYCENT, and from 548 to 17230 kg ha<sup>-1</sup> for DNDC. The regression of modelled versus measured yields had R<sup>2</sup> values of 0.60 and 0.66, bias estimates of -823 and -578 kg ha<sup>-1</sup>, and RMSE values of 2201 and 2088 kg ha<sup>-1</sup> for DAYCENT and DNDC, respectively (Table 2). Both models had a significant relationship with observed values based on different crop types (Fig. 1), but there were significant differences from the measured values according to t-tests (Table 2).

### 3.2 Daily N<sub>2</sub>O emission validation

Seasonal patterns of daily N<sub>2</sub>O emissions were analysed for 5 sites with latitudes between 28.6° to 47.4° and longitudes from 113.3° to 126.6° (Table 3), representing several different climate regions and most common cropping systems in China. Seasonal emission patterns simulated by the DNDC and DAYCENT models were generally similar to the observed values for most of the experimental period. Also, a significant increasing trend in N<sub>2</sub>O emissions was simulated with increasing N application rates, corresponding with experimental observations. Both DAYCENT and DNDC models failed to model the specific timing and magnitude of daily N<sub>2</sub>O emission peaks. Overall, the DNDC model overestimated emissions on days with high precipitation by a factor of around 2, particularly at the upland sites (Fig. 2c and 2d). The DAYCENT model overestimated the fluxes upon drainage of rice cultivation systems (Fig. 2a, 2b and 2e).

### 3.3 Cumulative N<sub>2</sub>O emission validation

The observed emissions from 425 field N<sub>2</sub>O emission measurements across 67 studies (Fig. 1) ranged from 0 to 11.14 kg N ha<sup>-1</sup> with N fertilizer applied in the range of 0-600 kg N ha<sup>-1</sup>.

The regression of modelled versus observed emissions had an  $R^2$  of 0.14 for both DAYCENT and DNDC model, 0.23 for LRM, 0.15 for IPCC (Table 1, Fig. 3). Moreover, the LRM had the lowest RMSE and bias with the values of 1.22 and -0.02 kg N ha<sup>-1</sup>, respectively; the DNDC model had the highest RMSE of 1.48 kg N ha<sup>-1</sup> and bias of -0.52 kg N ha<sup>-1</sup> (Table 1). According to the t-test, the DNDC model results were significantly different from the observed values, but estimated values were not significantly different from observations for the other three models (Table 1).

### 3.4 Model accuracy

We also assessed the impacts of the simulated N<sub>2</sub>O emissions for different fertilizer and crop types, and observed that the accuracy of the four models differed (Table 4). The DAYCENT model estimated N<sub>2</sub>O emissions from mineral and organic fertilizer types with the lowest RMSE and bias, and did not differ significantly from the measured values. Conversely, the IPCC significantly overestimated the emissions with organic fertilization but estimated N<sub>2</sub>O emissions with low RMSE and bias for mineral fertilizers. In fact, only the DNDC model significantly underestimated the emissions under the mineral fertilizer treatments. The estimated values from all models showed significant differences compared to measured values under the control treatment with no N inputs. For crop types (Table 5), the LRM performed well for rice and maize cultivated system, as did the IPCC method for maize, and the DAYCENT model for wheat. Moreover, all models simulated emissions for soybean well, but none performed particularly well for cotton and fallow.

N management, and particularly additions, are the most important drivers of soil N<sub>2</sub>O emissions (Del Grosso et al., 2009). Given this fact, we further compared the correlations of N addition rates with observed emission values for the models (Fig. 4; Table 6). Both the modelled and observed values had a similar response to fertilizer application rate. The modelled values were higher in the paddy rice system (Fig. 4a) and were lower in the upland

241 cropping system (Fig. 4b) compared to the measured values at low N application rates. The  
242 range of the slopes were 0.0018-0.0042 and 0.0039-0.0056 for paddy rice and upland  
243 cropping systems, respectively.

244

## 4 Discussion

Given the recognised difficulty in estimating N<sub>2</sub>O emissions precisely and the ongoing challenge of developing models which perform over a wide range of conditions, model inter-comparisons are an important way to determine a best candidate model for a given region and to identify potential ways to reduce the uncertainties. Model inter-comparisons have previously been carried out in several countries (Frolking et al., 1998; Smith et al., 2007; Abdalla et al., 2010).

Reasonable simulation of crop yield is of key importance to accurately predict N<sub>2</sub>O emissions for process-based models of plant-soil systems. The two process-based models, DAYCENT and DNDC, performed well in simulating crop yield, explaining 64% of the variation in observed yields with DAYCENT and 71% with DNDC (Table 2, Fig. 1). However, both models significantly underestimated yields by 823 and 578 kg ha<sup>-1</sup> for DAYCENT and DNDC, respectively, as indicated by the bias and t-test (Table 2). Previous studies have demonstrated reasonably accurate and precise predictions of crop yields in China for both DAYCENT and DNDC models (Cheng et al., 2013; Qiu & Wang, 2012). However, some studies suggested some bias in model simulations. For example, Cheng et al. (2014) found that the DAYCENT model underestimated corn yields by 521.59 kg ha<sup>-1</sup>, and Cui et al. (2014) found that the DNDC model underestimated the plant biomass for cotton. The bias in DNDC may be associated with the fact that DNDC does not simulate phosphorus and potassium impacts on production. In addition, the climate data used for the two process-based models includes only the maximum/minimum temperature and precipitation, which also might result in uncertainties for model simulation, and may be improved if other climate variables were addressed, such as the influence of humidity on transpiration rates and water stress. We found that the simulated yield varies greatly between control and fertilized plots for DAYCENT, which resulted in large bias compared with measured values. Production algorithms in

270 DAYCENT may be too sensitive to N availability. Sansoulet et al. (2014) also found that  
271 DAYCENT was less effective at predicting biomass under limited N rates compared to  
272 DNDC.

273 In general, the models were able to simulate the daily flux over time; however, there were  
274 some abnormal peak periods of emissions simulated by both models, compared to the  
275 observed emissions. Specifically, N<sub>2</sub>O emission peaks often appeared in simulated upland  
276 crops of maize and wheat after heavy rainfall events for DNDC (Fig. 2c-2e), indicating N<sub>2</sub>O  
277 emissions are highly sensitive to soil moisture dynamics in the models (Lessard et al., 1996;  
278 Frohking et al. 1998; Smith et al., 2002). In addition, Smith et al., (2008) observed that  
279 DAYCENT and DNDC models both had difficulty in capturing soil water content accurately.  
280 Soil moisture dynamics are linked to soil texture. Groffman and Tiedje (1989) suggested that  
281 the smaller average pore size in finer textured soils leads to greater soil water retention and  
282 greater opportunity to create anaerobiosis, while denitrification occurs at lower rates in a  
283 well-drained coarse-textured soil (Bouwman et al., 2002a, 2002b). Thus, there may be an  
284 opportunity to further resolve the relationship between soil texture and water-filled pore  
285 space, and improve model predictions. Also, the accuracy of capturing N<sub>2</sub>O emission peaks  
286 may be associated with the frequency of sampling, with low frequency sampling (e.g., once a  
287 week or month) missing some of the peaks that are captured by the models.

288 In general, the four models explained 14%~23% of the variation in observed seasonal  
289 cumulative N<sub>2</sub>O emissions. N<sub>2</sub>O emissions are inherently difficult to predict precisely for  
290 reasons stated above; however, this does suggest considerable opportunity for improvement.  
291 Nevertheless, no significant bias was identified except for the significant bias of -0.52 kg N  
292 ha<sup>-1</sup> for the DNDC model (Table 2). Beheydt et al. (2007) reported an overestimation of 7.4  
293 kg N<sub>2</sub>O-N ha<sup>-1</sup> for DNDC based on 22 long-term N<sub>2</sub>O field experiments. In addition, other  
294 research found that DAYCENT performed better than in this study, with an R-squared of 78%

which was much higher than the value found in this study (Cheng et al., 2014). In this study we used more field measurements than Cheng et al. (2014), which may have added heterogeneity and uncertainty in model simulation. Conversely, Abdalla et al. (2010) indicated that DAYCENT performed poorly when simulating control plots, with N<sub>2</sub>O flux of -57% below the measured values. Additionally, several studies have indicated that model accuracy varied for different fertilizer and cropping types (Smith et al., 2002; Cheng et al., 2014; Albanito et al., 2017). As shown, the DAYCENT model performs well with mineral and organic fertilizer types. The IPCC default method could only accurately predict emissions associated with mineral N fertilization, similar to results from Li et al. (2001).

DNDC did not accurately simulate N<sub>2</sub>O emissions associated with mineral fertilizer and paddy rice (Table 4, 5). In contrast, Smith et al. (2002) found the DNDC model prediction of N<sub>2</sub>O flux from control, manure, and mineral fertilization corresponded well with observed measurements from maize in Canada. Regardless, Li et al. (2017) reported that DNDC was not suitable for China as it lacks a number of features which are crucial for representing Chinese agro-ecosystems, especially paddy rice cultivation, complex and multiple cropping systems, and intensive management practices.

There are different target functions for the four models. The predictions of LRM and IPCC methods were more accurate and precise than the process-based models. While the LRM model was only used to calculate fertilizer-induced N<sub>2</sub>O emissions based on the underlying datasets that were used to derive these functions (and therefore not independent data), this does indicate that if a reasonably comprehensive dataset of N<sub>2</sub>O emissions exists for a given region, then better predictions will be obtained from a linear regression model than by calibrating and deploying a process-based model. The two process-based models, in theory, should be able to capture more heterogeneity and be applied across a broader range of croplands in China. One of the key strengths of DAYCENT is the initialization of SOM pools

to accurately represent the carbon stocks, and the linkage between C and N flows through the plant-soil system. The N associated with carbon lost in respiration (30% to 80% of the carbon flow is respired) is mineralized and becomes substrate of nitrification and denitrification (DAYCENT user manual). DNDC also has strengths related to fertilizer applications at varying depth, and a more mechanistic representation of N dynamics with Michaelis-Menten dynamics (Li et al., 2006).

Process-based models, such as DAYCENT and DNDC, can also represent more management impacts than empirical functions, particularly if data are limited for fitting a statistical model. For example, Xu et al. (2000) showed a significant effect of splitting fertilizer into three or more applications in DNDC, reducing N<sub>2</sub>O emissions by 25%. Field practices of irrigation and tillage also influence N<sub>2</sub>O fluxes, and their impacts can be represented in these simulation models.

Our results indicated that the accuracy of model simulations may differ across a range of N rates. Cheng et al. (2014) showed DAYCENT tended to underestimate N<sub>2</sub>O emissions at higher measured emission rates, which were also seen for paddy rice in Fig. 4a. Albanito et al. (2017) studied N<sub>2</sub>O-EFs and found that they tended to decrease with the N application rates approaching 1% in crops fertilized above 300 kg N ha<sup>-1</sup>, and the IPCC-EF would tend to underestimate N<sub>2</sub>O emissions by approximately 21% below a fertilization of 200 kg N ha<sup>-1</sup>. Similarly, Shcherbak et al. (2014) indicated that the IPCC-EF would underestimate and overestimate N<sub>2</sub>O emissions in croplands fertilized above and below the threshold of approximately 150 kg N ha<sup>-1</sup>. Sansoulet et al. (2014) also showed the different sensitivity under limited and high N rates. The negative intercept for DNDC might indicate that emissions are under-estimated with no fertilizer applied.

Environmental factors (especially climate) and human-induced activities (e.g. fertilizer, tillage, straw return, irrigation) influence N<sub>2</sub>O producing processes over both temporal and

345 spatial scales, resulting in heterogeneous N<sub>2</sub>O emissions at field level (Flessa et al., 2002).  
346 Cumulative seasonal N<sub>2</sub>O emissions based on the closed static chamber method were used in  
347 most of the experiments at monthly or weekly intervals, which may lead to high inherent  
348 variability of N<sub>2</sub>O fluxes. Ju et al. (2011) showed that a sampling frequency of 3 or 6 days led  
349 to 112-236% overestimation of total N<sub>2</sub>O emissions. Process-based models may predict a flux  
350 peak during times, such as after a rainfall event, which is not represented in observational  
351 datasets with a low sampling frequency. Hence, an overestimation or underestimation of  
352 N<sub>2</sub>O fluxes from upland soils may occur with static chambers, and more continuous  
353 measurements will likely reduce uncertainties in evaluating models (Yao et al. 2009; Ju et al.  
354 2011).



## 5 Conclusion

The performance of the four models varied for the cropping systems and fertilization management practices. Consequently, we conclude that neither of the models emerged as a clear “best” choice for estimating N<sub>2</sub>O emissions for Chinese cropping systems. In the short term, it may be best to adopt the methods based on linear regression models to calculate the N<sub>2</sub>O emissions for rice, maize, wheat and soybeans, although even this approach has limitations, leading to significant differences between observed and modelled emissions for cotton, fallow or rape. Further development is needed to represent regional conditions in China associated with dominant soil properties, agricultural practices, cropping systems, and climate conditions, in order to refine empirical models and improve the suitability of process-based models in Chinese conditions.

## Acknowledgments

This work was financially supported by China Natural Science Foundation under a grant number 41501569 and “the Fundamental Research Funds for the Central Universities” under a grant number KJQN201673. This work was also supported by Department of Science and Technology of Jiangsu province under a grant number BK20150684. This work also contributes to the activities of NCircle - a BBSRC-Newton Funded project (BB/N013484/1). The first author thanks the China Scholarship Council (CSC) for funding to support study at University of Aberdeen, UK.

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468

469    **Table captions**

470    Table 1 Models inputs for models simulation.

471    Table 2 Statistics describing the performance of DAYCENT and DNDC model in grain yield  
472    simulations.

473    Table 3 Information of sites selected for simulating daily fluxes of N<sub>2</sub>O emissions.

474    Table 4 The performance of four models in estimating N<sub>2</sub>O emissions under different  
475    fertilizer management

476    Table 5 The performance of four models for estimating N<sub>2</sub>O emissions associated with crop  
477    types

478    Table 6 Statistics describing the correlations of observed or simulated N<sub>2</sub>O emissions with  
479    nitrogen fertilizer application rates in Fig. 4.

480

481 **Figure captions**

482 Fig.1 Comparison of measured and simulated crop yields for experimental sites across China

483 Fig. 2 Comparison of observed and modeled daily patterns of N<sub>2</sub>O emissions from rice paddy  
484 sites. (The representation of letters “a” to “e” were explained in Table 3)

485 Fig. 3 Comparison of observed and simulated cumulative N<sub>2</sub>O emissions for four models

486 Fig. 4 Comparison of observed and modelled growing season N<sub>2</sub>O emissions from a range of  
487 nitrogen fertilizer application rates (a, rice paddy; b, upland)



Table 1 Models input data for models simulation.

Data items	DAYCENT	DNDC	LRM	IPCC
Geographical location	Longitude; Latitude;	Latitude	/	/
Climate factors	Daily maximum temperature; daily minimum temperature; daily precipitation; N decomposition;	The same as for DAYCENT	Annual average temperature	/
Soil properties	SOC; C/N ratio; soil clay, silt, sand content; soil bulk density; pH;	The same as for DAYCENT	Soil clay content	/
Growing time	Crop type; sowing date; harvested date;	The same as for DAYCENT	Crop type	/
Management practices	N applied rate, date and type; irrigating amount and date; tillage intensity and date; straw returning amount	The same as for DAYCENT	N rate; N fertilizer type	N rate

“/”: The parameters were not required to be entered.

Table 2 Statistics describing the performance of DAYCENT and DNDC model in grain yield simulations.

Items	Models	R-square	RMSE	Bias	t-test
Yield	DAYCENT	0.64	2201	-823	s
	DNDC	0.71	2088	-578	s
N <sub>2</sub> O-N	DAYCENT	0.14	1.35	-0.15	ns
	DNDC	0.14	1.48	-0.52	s
	LRM	0.23	1.22	-0.02	ns
	IPCC	0.15	1.42	-0.09	ns

Table 3 Information of sites selected for simulating daily fluxes of N<sub>2</sub>O emissions.

Site	Latitude, Longitude	Region	Cropping system	Typical N fertilizer rate (kg ha <sup>-1</sup> )	References
Heilongjiang (a)	47.4°,126.6°	Northeast China	RP	95.4	Yue et al., 2005
Hunan (b)	28.6°,113.3°	South-Central China	RP-RP	135(RP), 135(RP)	Wang et al., 2014
Liaoning (c)	41.8°,123.6°	Northeast China	MA	160	Cheng et al., 2016
Hebei (d)	40.0°,118.1°	North China	MA	180	Lu et al., 2015
Jiangsu (e)	32.0°,118.8°	East China	RP-WH	250(RP), 250(WH)	Zhou et al., 2016

Table 4 The performance of four models in estimating N<sub>2</sub>O emissions under different fertilizer management

Fertilizer type	Model	RMSE	Bias	t-test
No fertilizer	DAYCENT	0.81	-0.21	s
	DNDC	0.87	-0.51	s
	LRM	0.73	0.16	s
Mineral fertilizer	DAYCENT	1.52	-0.06	ns
	DNDC	1.71	-0.66	s
	LRM	1.37	-0.16	ns
	IPCC	1.50	-0.07	ns
Organic fertilizer	DAYCENT	0.53	0.01	ns
	DNDC	0.54	-0.15	ns
	LRM	0.77	0.41	s
	IPCC	1.65	1.40	s

Table 5 The performance of four models for estimating N<sub>2</sub>O emissions associated with crop types

Crop type	Model	RMSE	Bias	t-test	Crop type	Model	RMSE	Bias	t-test
Rice	DAYCENT	1.02	0.11	ns	Cotton	DAYCENT	3.14	-3.10	s
	DNDC	1.00	-0.41	s		DNDC	4.43	-4.25	s
	LRM	0.93	-0.04	ns		LRM	3.54	-3.39	s
	IPCC	0.94	-0.34	s		IPCC	2.34	-2.34	ns
Maize	DAYCENT	1.68	-0.32	ns	Fallow	DAYCENT	1.46	-1.09	s
	DNDC	1.72	-0.51	s		DNDC	1.51	-1.26	s
	LRM	1.38	-0.16	ns		LRM	1.21	-0.73	s
	IPCC	1.64	0.05	ns		IPCC	2.04	-1.72	s
Wheat	DAYCENT	1.19	-0.06	ns	Rape	DAYCENT	0.69	0.35	ns
	DNDC	1.33	-0.55	s		DNDC	1.54	-1.33	s
	LRM	1.29	0.48	s		LRM	0.86	0.73	s
	IPCC	1.56	0.52	s		IPCC	0.46	-0.09	ns
Soybean	DAYCENT	0.89	-0.44	ns					
	DNDC	0.59	-0.13	ns					
	LRM	0.99	-0.58	ns					
	IPCC	0.85	-0.59	ns					

Table 6 Statistics describing the correlations of observed or simulated N<sub>2</sub>O emissions with nitrogen fertilizer application rates in Figure 4.

	Paddy rice			Upland crop		
	Slope	intercept	R-square	Slope	intercept	R-square
Observed	0.0034	0.3180	0.1250	0.0042	0.8404	0.1174
DAYCENT	0.0042	0.3270	0.2383	0.0046	0.4888	0.3872
DNDC	0.0027	-0.0243	0.3418	0.0056	0.0561	0.3316
LRM	0.0018	0.4722	0.4522	0.0039	0.8809	0.4463

Fig.1  
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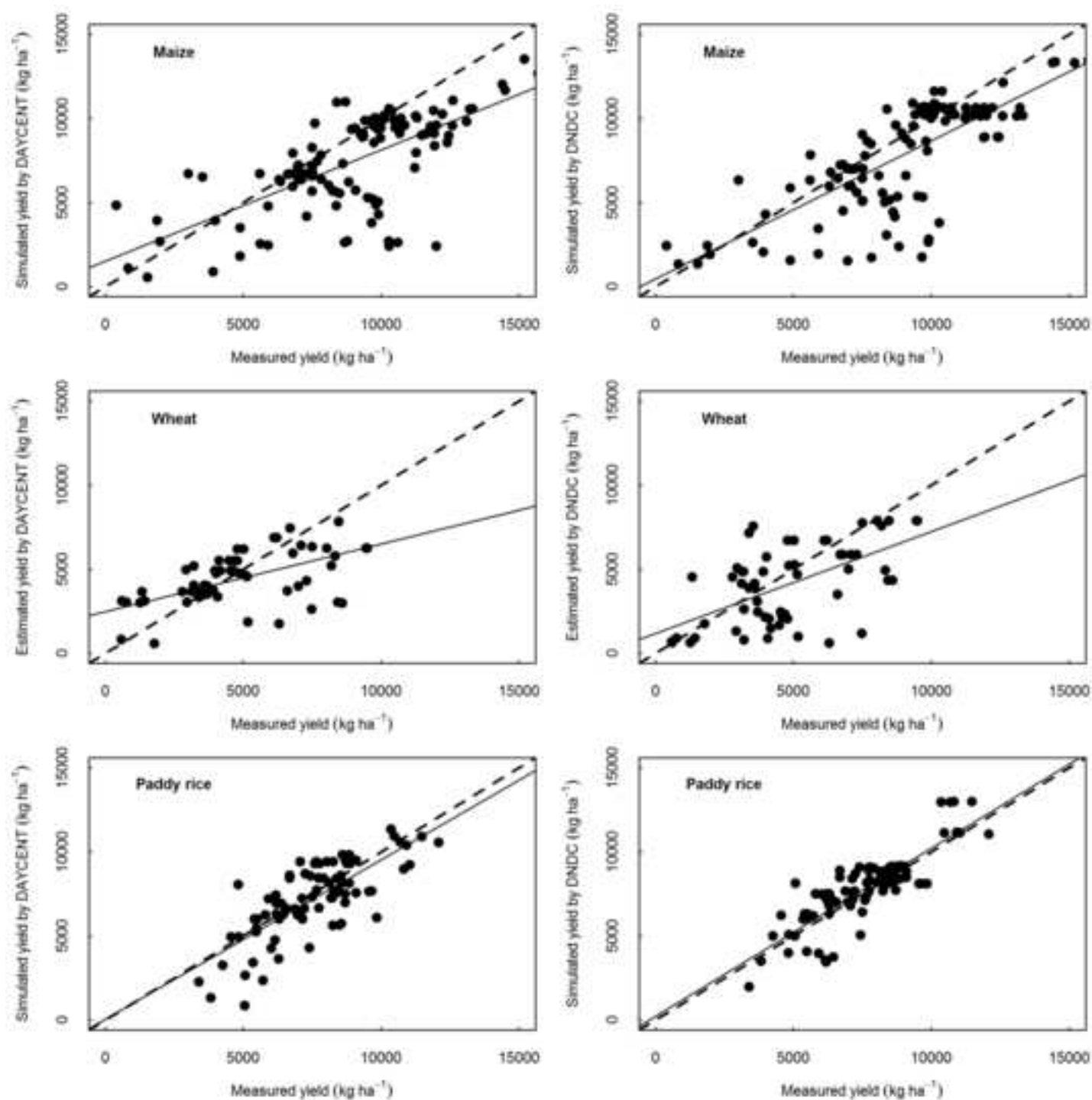


Fig.2

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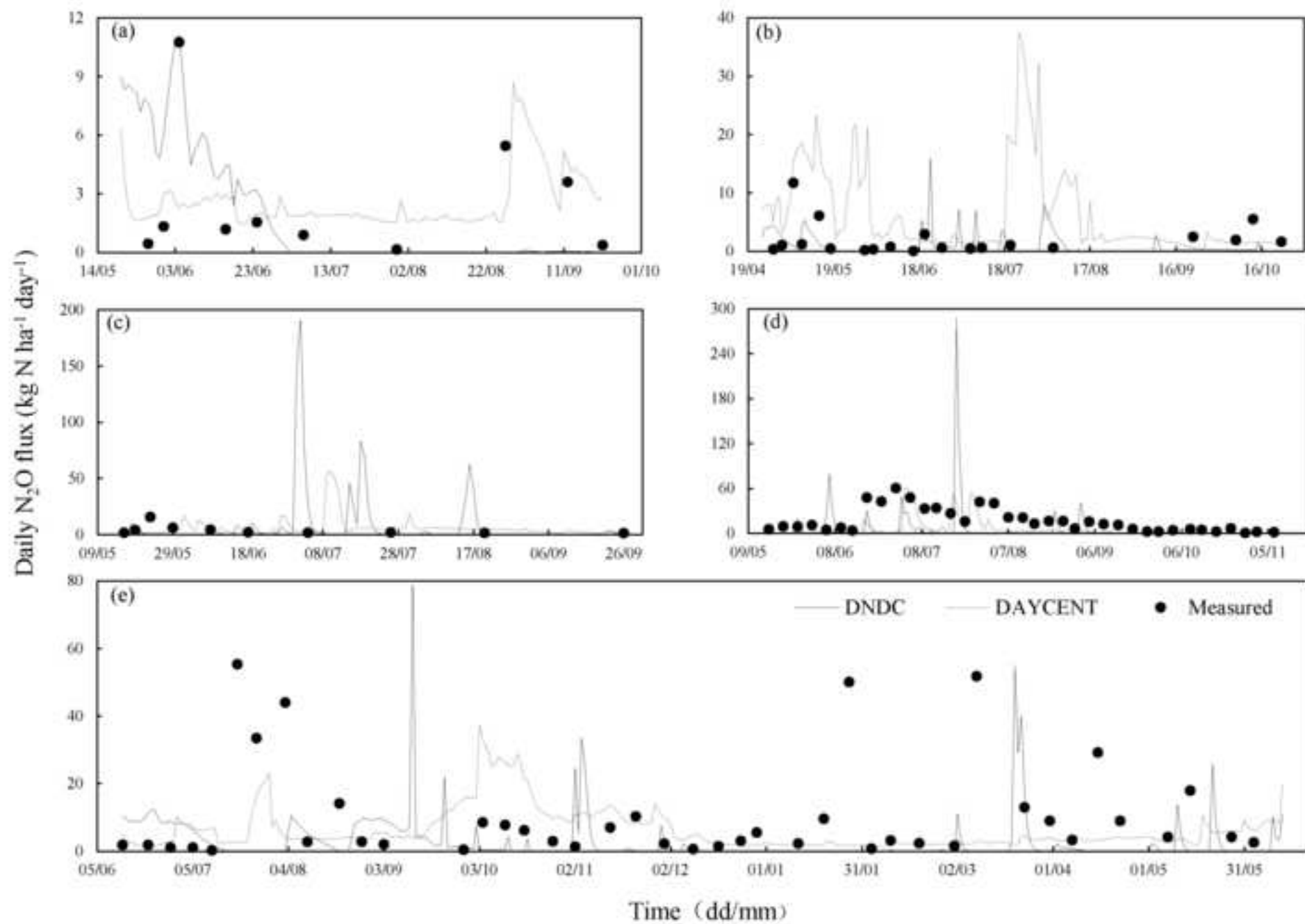
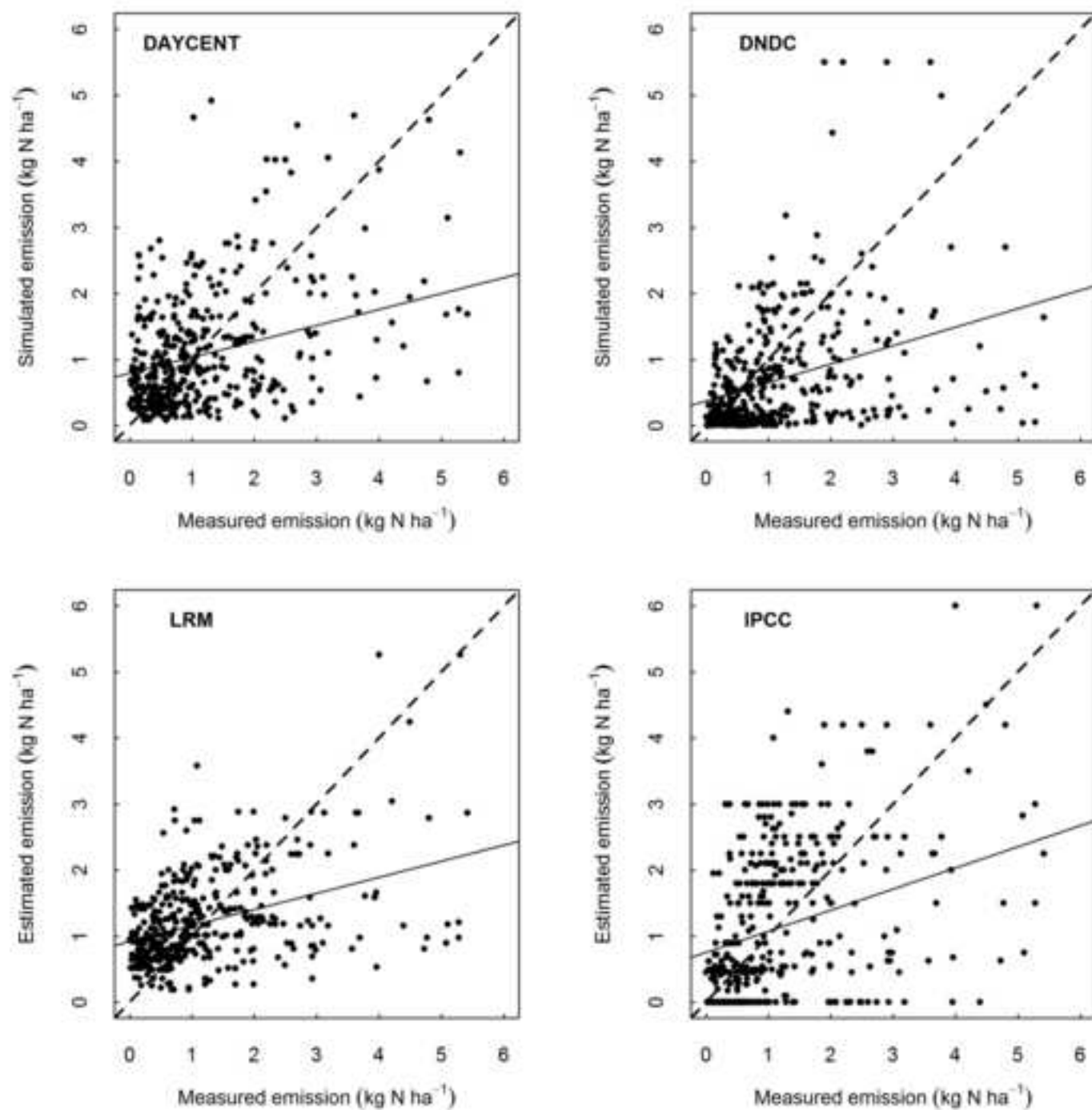




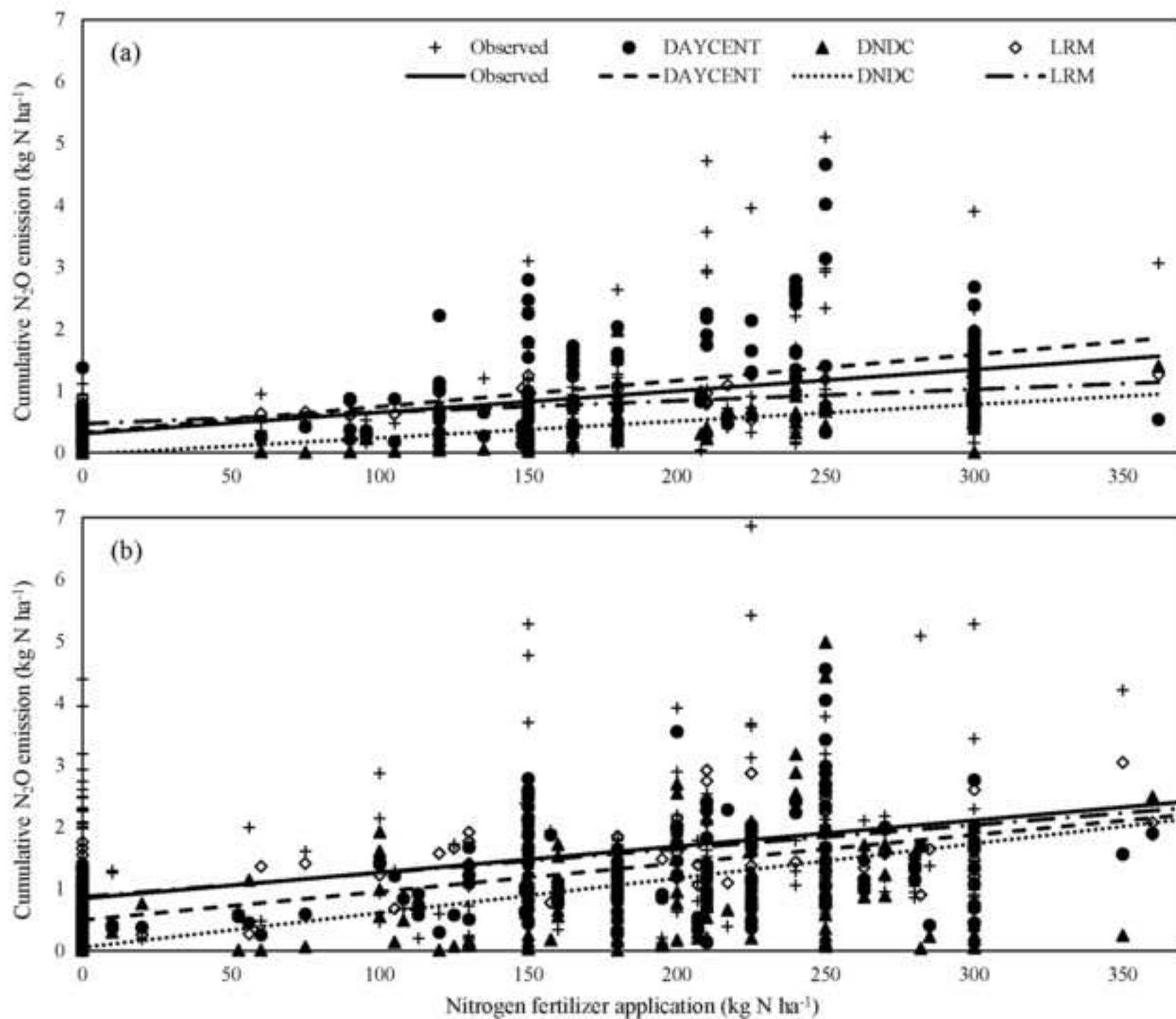
Fig.3

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**Fig.4**

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**Table S1**

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